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# **Temperature-Compensated High-Resolution Infrared Absorption Spectroscopy: Application to Carbon Monoxide**

C. J. Fisher and W. P. Dixon  
ARO, Inc.

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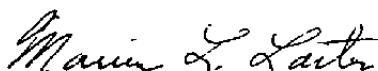
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20 ABSTRACT (Continue on reverse side if necessary and identify by block number) <p>The analytic basis of an optical absorption technique suitable for a single line-of-sight determination of average gaseous species number densities in the presence of concentration, pressure and temperature gradients, and particulate loading is presented and applied to carbon monoxide (CO). Applying the technique involves using the power of very high-resolution infrared (IR) spectroscopy, now made available by tunable diode</p>		

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### 20. ABSTRACT (Continued)

lasers. The method requires the construction of functions of molecular line strengths that are nearly temperature independent. A method of constructing these functions is described and applied to the case of CO. Possible applications to practical combustion systems are also outlined.

## **PREFACE**

The work reported herein was conducted by the Arnold Engineering Development Center (AEDC), Air Force Systems Command (AFSC). The results of the research were obtained by ARO, Inc., AEDC Group (a Sverdrup Corporation Company), operating contractor for the AEDC, AFSC, Arnold Air Force Station, Tennessee, under ARO Project Number P32M-12. The Air Force project manager was Dr. H. E. Scott (DOT). The data analysis was completed on June 15, 1979, and the manuscript was submitted for publication on November 29, 1979. The authors wish to express their appreciation to Mr. K. R. Kneile and Mr. R. D. Meyer, ARO, Inc., for their work in numerical analysis and computer programming.

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## 1.0 INTRODUCTION

Practical combustion system gas flows are normally characterized by temperature, concentration, and pressure gradients, and, in some cases, significant particulate loadings. The combustion diagnostician must often provide spatially resolved and/or average concentration determinations of various gaseous components of the discharge. This report presents the basis for developing an optical absorption technique suitable for a single line-of-sight determination of average gaseous species number density (moles/volume) in the presence of concentration, pressure and temperature gradients, and particulate loadings. The technique is dependent on the power of very-high-resolution spectroscopy (i.e., its application requires detailed knowledge of absorption line shapes free of instrumental degradation). Such capabilities have been made available with tunable diode laser spectroscopy.

Techniques for providing average species number density determinations rely on spatially resolved measurement followed by appropriate averaging. These techniques include probe measurements, scanned Raman and fluorescence measurements, and scanned absorption and/or emission measurements followed by an inversion procedure. These techniques all involve either spatial scanning and/or multiple detection arrangements. In many applications a single average species number density determination, rather than a profile, is satisfactory. A single line-of-sight absorption measurement of average species number density is possible in the presence of concentration, temperature, and pressure gradients and particulate loadings if the following conditions are met:

1. The source beam is passed through the environment of interest in the form of a collimated beam of sufficient width to adequately sample the volume to be averaged.
2. The absorption data are reducible to a function only of number density of the species of interest and of the path length, and are thus independent of temperature, pressure, and beam attenuation attributable to particulates or optical component degradation.

This report develops the basis for constructing functions of very-high-resolution absorption data that have these desired properties.



## 2.0 SOME PRINCIPLES OF QUANTITATIVE VERY-HIGH-RESOLUTION SPECTROSCOPY

The most direct route to quantitative spectroscopic information from absorption measurements is the line strength,  $S$ , defined for a single absorption line as

$$S = \int_{-\infty}^{+\infty} k(\nu) d\nu \quad (1)$$

where  $k(\nu)$  is the absorption coefficient. The absorption coefficient is defined for a homogeneous path by the relationship

$$\tau(\nu) = \exp[-k(\nu)\ell] \quad (2)$$

where  $\tau(\nu)$  is the transmittance at wavelength  $\nu$ , and  $\ell$  is the path length. It can be shown (Ref. 1) that  $S$  is a function of only the temperature, number density, and properties of the isolated molecule. Specifically,  $S$  is given by

$$S = \int_{-\infty}^{+\infty} k(\nu) d\nu = \frac{\pi e^2}{m_e c^2} N_\ell f \left( 1 - \frac{g_\ell}{g_u} \frac{N_u}{N_\ell} \right) \quad (3)$$

where

- $e$  is the charge of an electron,
- $m_e$  is the mass of an electron,
- $c$  is the speed of light,
- $f$  is the oscillator strength, a measure of the relative strength of the transition,
- $N_\ell, N_u$  are, respectively, the lower- and upper-state number densities, and
- $g_\ell, g_u$  are, respectively, the lower- and upper-state degeneracies.

In contrast to the equivalent width,  $EW$ , of the absorption line, the line strength is independent of broadening. The equivalent width of a line is given by

$$EW = \int_{-\infty}^{+\infty} [1 - \tau(\nu)] d\nu \quad (4)$$

The temperature dependency of  $S$  is due solely to the change in number density with temperature of the lower and upper states, primarily the lower state. The line strengths of lines originating from different lower states have different temperature dependencies. The number density of a state "i" is given by

$$N_i = N_T \frac{g_i \exp(-E_i/kT)}{Q(T)} \quad (5)$$

where

$N_T$  is the total number density of the species of interest.

$E_i$  is the energy of state i,

$g_i$  is the degeneracy of state i,

$T$  is the temperature (K),

$k$  is the Boltzmann constant, and

$Q(T)$  is the internal partition function, a known function of the molecular constants.

Following the substitution of Eq. (5) into Eq. (3), the line strength of an individual line as a function of temperature is found to be

$$S(T) = \frac{\pi e^2 f}{m_e c^2} N_T \frac{g_l \exp(-E_l/kT)}{Q(T)} \left\{ 1 - \exp \left[ (E_l - E_u)/kT \right] \right\} \quad (6)$$

where  $E_u$  and  $E_l$  are, respectively, the upper and lower energy levels.

In terms of a tabulated line strength per molecule at a reference temperature,  $S_m(T_o)$  ( $\text{cm mol}^{-1}$ ),  $S(T)$  is given by

$$S(T) = S_m(T_o) \frac{N_l \exp(-E_l/kT) Q(T_o) \left\{ 1 - \exp \left[ (E_l - E_u)/kT \right] \right\}}{\exp(-E_l/kT_o) Q(T) \left\{ 1 - \exp \left[ (E_l - E_u)/kT_o \right] \right\}} \quad (7)$$

If the molecular constants required for the number density calculation, the line strength at a standard temperature and pressure, the temperature, and the path length are known, then the number density of the infrared-active species of interest can be calculated from a single absorption line (i.e., without knowledge of collisional broadening effects) provided that the following conditions are met:

1. The line is not saturated. If the line is saturated, the absorption coefficient cannot be calculated near the center of the line with use of Eq. (2). This limitation should not be serious, inasmuch as a weaker nearby line should be easy to find for most molecules.
2. All of the absorption line is observable. For broad lines wing absorption is often significant but not observable because of the presence of noise and/or overlapping lines. If this is the case, modeling of the line is required to correct for the unobserved wings.

The problem of calculating number densities from absorption spectra is greatly simplified by using a very-high-resolution instrument (i.e., one that is capable of producing true line shapes without instrumental distortion). With the accurate absorption spectra, the crucial transformation from transmission to absorption coefficient can be made with Eq. (2), and the line strength can be calculated directly. Without this high resolution, the equivalent width method must be employed. In addition to the information needed for the line strength method, this method requires broadening coefficients at the temperatures of interest for all species present. This information is generally not currently available for elevated temperatures.

### 3.0 CONSTRUCTION OF GRADIENT-INDEPENDENT FUNCTIONS

As indicated in Eq. (3), the integral of the absorption coefficient—the line strength—is a function of only the number density of the species of interest and the temperature. The independence of the line strength with respect to beam obstructions such as particulates is a consequence of the nature of the transmittance measurement. The transmittance is the ratio of the radiation intensity at the detector with the species of interest in the optical path to the intensity without the species. Thus, any obstructions in the path reduce the two intensities by the same fraction and do not change the ratio. The independence of the line strength with respect to pressure and, hence, pressure gradients is a simple consequence of the independence of the line strength with respect to line shape phenomena such as the pressure-broadening effects indicated in Eq. (3).

Thus, the line strength,  $S$ , of a single, resolved, nonsaturated absorption line, obtainable directly from analysis of very-high-resolution spectra, meets all the criteria for single line-of-sight average number density measurement technique except for the temperature dependency of the line strength. This last obstacle can be overcome with use of a function of two or more line strengths that is characterized by a negligible temperature dependency.

Conventional spectral notation (Ref. 3) is used in this report. An individual line is identified by the symbol  $v_u-v_l X (J_l)$  where  $v_u$  and  $v_l$  are, respectively, the upper- and lower-state vibrational quantum numbers,  $X$  is the rotational transition-type letter P or R, and  $J_l$  is the lower-state rotational quantum number.

As noted before, the line strengths of lines originating from different lower energy levels have different temperature dependencies. Figure 1 shows a few examples of some typical CO line strength,  $S_m$ , temperature dependencies calculated with Eq. (7).

With the exception of the 1-0 R(0) line, line strengths increase with temperature up to a temperature characteristic primarily of the lower-state energy level; at that point, they decrease. The higher the lower level, the higher the peak temperature. Thus, for any given temperature range, some line strengths will be monotonically increasing, some monotonically decreasing, and some peaking.

To devise nearly temperature-independent functions of line strengths, a linear combination was constructed of line strengths of differing temperature dependency. This linear combination is characterized by the least possible degree of temperature dependency over the temperature range of interest. The temperature-minimized function is defined as

$$F(T) = \sum_{i=1}^n a_i S_{mi}(T) \quad (8)$$

where

$F(T)$  is the function,

$S_{mi}(T)$  is the  $i$ th line strength (molecular units),

$a_i$  is the linear combination weighting coefficient of  $S_{mi}(T)$ , and

$n$  is the number of lines selected.

The  $a_i$  coefficients are chosen to minimize the temperature dependency of  $F(T)$  and are normalized so that  $F(T)$  averages to unity over the temperature range of interest. The temperature dependencies of the  $S_{mi}$ 's is calculated exactly with use of Eq. (7). The input CO  $S_m$ 's at a reference temperature (298.15 K) were calculated from the approximate formula of Herman and Wallis (Ref. 2) as used by Hanson (Ref. 4):

$$S_{m_{\substack{J_l \rightarrow J_l \pm 1 \\ v_l \rightarrow v_l + 1}}}(298.15) = 1.016 \times 10^{-7} \frac{\nu}{\nu} v_u \exp(-E_{v_u}/298.15k) \times \left[ (1 - \exp(-h\nu_c/298.15k)) S^J Q(298.15) \right] \quad (9)$$

where

- $\nu$  is the line wave number,
- $\bar{\nu}$  is the band center wave number,
- $E_l$  is the lower energy level,
- $k$  is Boltzmann's constant,
- $h$  is Planck's constant,
- $c$  is the speed of light,
- $S^J$  is equal to  $J_l$  for P lines and equal to  $J_l + 1$  for R lines, and

$Q(298.15)$  is the internal partition function at 298.15 (108.19).

The 1-0 band strength at 298 K of  $1.016 \times 10^{-7}$  cm mol $^{-1}$  of Varanasi and Sarangi (Ref. 5) was used. It should be noted that changes in the input  $S_m(298.15)$ 's would change only the calculated  $a_i$ 's, and not the temperature dependency of  $F(T)$ .

The procedure used to construct  $F(T)$  is as follows:

1. The temperature range of interest is chosen.
2. A set of  $n$  lines with differing temperature dependencies over that range is selected. (Other desirable criteria for line selection are discussed below.)
3. The  $p$  temperature  $T_j$ 's in the range of interest at which the function  $F(T) - 1$  is to be minimized are selected.
4. The  $p$  linear equations to be solved, one for each  $T_j$ , are constructed:

$$0 = \left[ \sum_{i=1}^n a_i S_{m_i}(T_j) \right] - 1 \quad (10)$$

5. The  $p$  equations for the  $n$  unknowns, the  $a_i$ 's, are solved. If  $p = n$ , then this system of equations can be solved exactly, provided that all the lines chosen have different temperature dependencies. If  $p > n$ —and this is the situation for all cases reported herein—then there are more equations than unknowns and, hence, no exact solution. This set of redundant equations was solved by an IBM System

370 computer using a least-squares procedure; the computer solved for a set of  $a_i$ 's that would minimize the sum of the squares of the deviations of  $F(T)$  from unity. Thus, the function  $G$  is minimized with respect to all the  $a_i$ 's:

$$G = \sum_{j=1}^p \left\{ \left[ \sum_{i=1}^n a_i S_{mi}(T_j) \right] - 1 \right\}^2 \quad (11)$$

Several criteria were used in the selection of the lines and in the evaluation of the resultant  $F(T)$ 's:

1.  $F(T)$  does not differ from unity by more than  $\pm 10$  percent at any temperature in the range of interest. Adding more lines to the function or restricting the temperature range generally results in a higher degree of temperature independence.
2. There are no interfering lines of  $\text{CO}$ ,  $\text{CO}_2$ , or  $\text{H}_2\text{O}$  strong enough to interfere with measurements of the selected lines within  $\pm 0.3 \text{ cm}^{-1}$  of the selected lines as indicated in the AFGL Atmospheric Absorption Line Parameters Compilation Tape (Ref. 6).
3. To facilitate rapid data acquisition, it is desirable to choose lines relatively close to each other in frequency ( $\pm 10 \text{ cm}^{-1}$ ).
4. Another means of facilitating rapid data acquisition is the use of the smallest possible number of lines. Only two- and three-line combinations were considered.
5. It is desirable for all of the  $a_i$ 's to be positive. This factor helps to lower uncertainties in the determination of number densities. (This point is discussed in a later section.)

## 4.0 RESULTS

Many combinations of lines meeting some or all of the above criteria were tested. The temperature range over which  $F(T) - 1$  was most often minimized was the 300- to 2000-K range, which represents the range of interest for the majority of possible combustion applications.

The minimized  $F(T)$ 's for 11 typical successful calculations are given in Figs. 2 through 12. The results of these calculations are summarized in Table 1. The best  $F(T)$ 's over the 300- to 2000-K range for closely spaced pairs of lines were obtained with the 1-0 R(12) line and either the 2-1 R(10, 20, or 21) lines (Figs. 2 through 4). Other good  $F(T)$ 's over the 300- to 2000-K range for pairs are the 1-0 P(10), 2-1 P(6) and the 1-0 P(11), 2-1 P(5) combinations (Figs. 7 and 8).

Many three-line sets yielded better than  $\pm 5$  percent temperature independence over the 300- to 2000-K range. A sample of these is presented in Figs. 9 through 12.

### 5.0 THE USE OF $F(T)$ IN CO NUMBER DENSITY DETERMINATIONS AND THE EFFECT OF EXPERIMENTAL UNCERTAINTIES

The procedure for determining the average CO number density is as follows:

1. The temperature range of interest and the set of lines to be used are selected and the  $a_i$ 's are solved for in the manner previously described.
2. The absorption spectra of the lines are recorded and the line strengths are calculated. If any of the lines is saturated, then all the line strengths are not obtainable, and weaker lines should be selected.
3. The average number density,  $\bar{N}_{CO}$ , is given simply by

$$\bar{N}_{CO} = F \quad (12)$$

where  $F$ , the linear combination of line strengths defined by Eq. (8), is evaluated with the experimentally evaluated line strengths ( $\text{cm}^{-2}$ ).

The range of CO concentrations detectable with the technique is large. There is virtually no maximum concentration because the very-low-abundance, isotopic species such as  $C^{13}O^{16}$ ,  $C^{12}O^{18}$ , and  $C^{13}O^{18}$  (i.e., unsaturated absorption lines, necessary for applying this technique) are always present. The greatest sensitivity is attained with long path lengths obtainable with multiple-pass optics. If there exists a 10-meter path length at 1 atm pressure and at 300 K, then a mole fraction of 2 ppm CO should be detected.

If only positive  $a_i$ 's can be found for  $F(T)$ , then the relative uncertainty of the CO number density is approximately the same as the relative uncertainty of the experimental line

strength determinations. The relative uncertainty of the CO determination,  $\Delta \bar{N}_{CO} / \bar{N}_{CO}$ , attributable to the experimental uncertainties in the  $S_i$ 's,  $\Delta S_i$ , is found—with use of the standard propagation of error treatment—to be

$$\frac{\Delta \bar{N}_{CO}}{\bar{N}_{CO}} = \sum_{i=1}^n |a_i| \frac{\Delta S_i}{S_i} \quad (13)$$

If the  $a_i$ 's differ in sign and are approximately equal in absolute value, then  $\Delta \bar{N}_{CO} / \bar{N}_{CO}$  becomes much greater than the  $\Delta S_i / S_i$ 's. This situation arises when lines characterized by similar temperature dependencies in the range of interest are selected. The increases in the experimental uncertainties arise from the fact that the calculation of  $\bar{N}_{CO}$  is dependent on the difference between large quantities, yielding a relatively small quantity. Therefore, the uncertainty of the large individual terms of  $F(T)$  can be quite large with respect to  $F(T)$ .

## 6.0 APPLICATIONS OF THE TECHNIQUE TO COMBUSTION FLOWS

The application of the high-resolution absorption temperature-compensated technique to combustion flow diagnostics is straightforward in principle. If the spectrometer source beam is expanded and collimated so that the beam-combustion flow overlap volume represents the entire flow, then the average number density in the flow can be obtained by using Eq. (12). The path length used in reducing the transmittance measurements [Eq. (2)] is the average length of the overlap volume along the direction of the collimated beam. A schematic of this application is shown in Fig. 13. The beam must be collimated rather than focused to prevent undue weight being placed on the average number density determination by a possibly atypical number density located at or near a focus.

For radially symmetric flows, a narrow beam can be used to generate species number density profiles. The information required for a spatial radial inversion consists of  $n$  line-of-sight transmittance measurements starting with a near-tangent measurement and progressing to the centerline. If the initial line of sight is assumed to intersect only one homogeneous ring to the line of sight, then the species number density of each ring can be determined by the temperature-compensated absorption technique, provided that none of the lines is saturated.

The transmittances of a sample consisting of  $n$  zones characterized by individual transmittances,  $\tau_i$ , is given by

$$\tau(\nu) = \prod_{i=1}^n \tau_i(\nu) \quad (14)$$



Substituting Eq. (2) into Eq. (14) and taking the logarithm of both sides yield

$$\ell_n \tau(\nu) = \sum_{i=1}^n k_i(\nu) \ell_i \quad (15)$$

where  $k_i$  and  $\ell_i$  are, respectively, the absorption coefficient and path length of zone  $i$ . By using Eq. (15) and the transmittance of the  $n$  lines of sight, one can determine the absorption coefficients as a function of wavelength and, hence, the line strengths of each zone. The species number densities can then be determined for each zone by using the temperature-compensated high-resolution technique. [It should be noted that the temperature of each zone could also be determined by ratioing the experimentally determined line strengths of lines originating from ground states of sufficiently different energies, as has been reported for homogeneous flat flames (Ref. 7).]

## 7.0 SUMMARY

The analytic basis of an optical absorption technique suitable for a single line-of-sight determination of average gaseous species number densities in the presence of concentration, pressure and temperature gradients, and particulate loading is presented and applied to CO. Applying the technique involves using the power of very-high-resolution IR spectroscopy, now made available by tunable diode laser spectroscopy. The method requires the construction of functions of molecular line strengths characterized by a minimal temperature dependency over large temperature ranges. A method of constructing these functions is described and applied to the case of CO. Eleven nearly temperature-independent functions of CO line strengths are presented. They are characterized by deviations from temperature independence of less than  $\pm 10$  percent over the temperature ranges from 300 to 2000 K and from 400 to 2000 K. Possible applications of the technique to practical combustion systems are also outlined.

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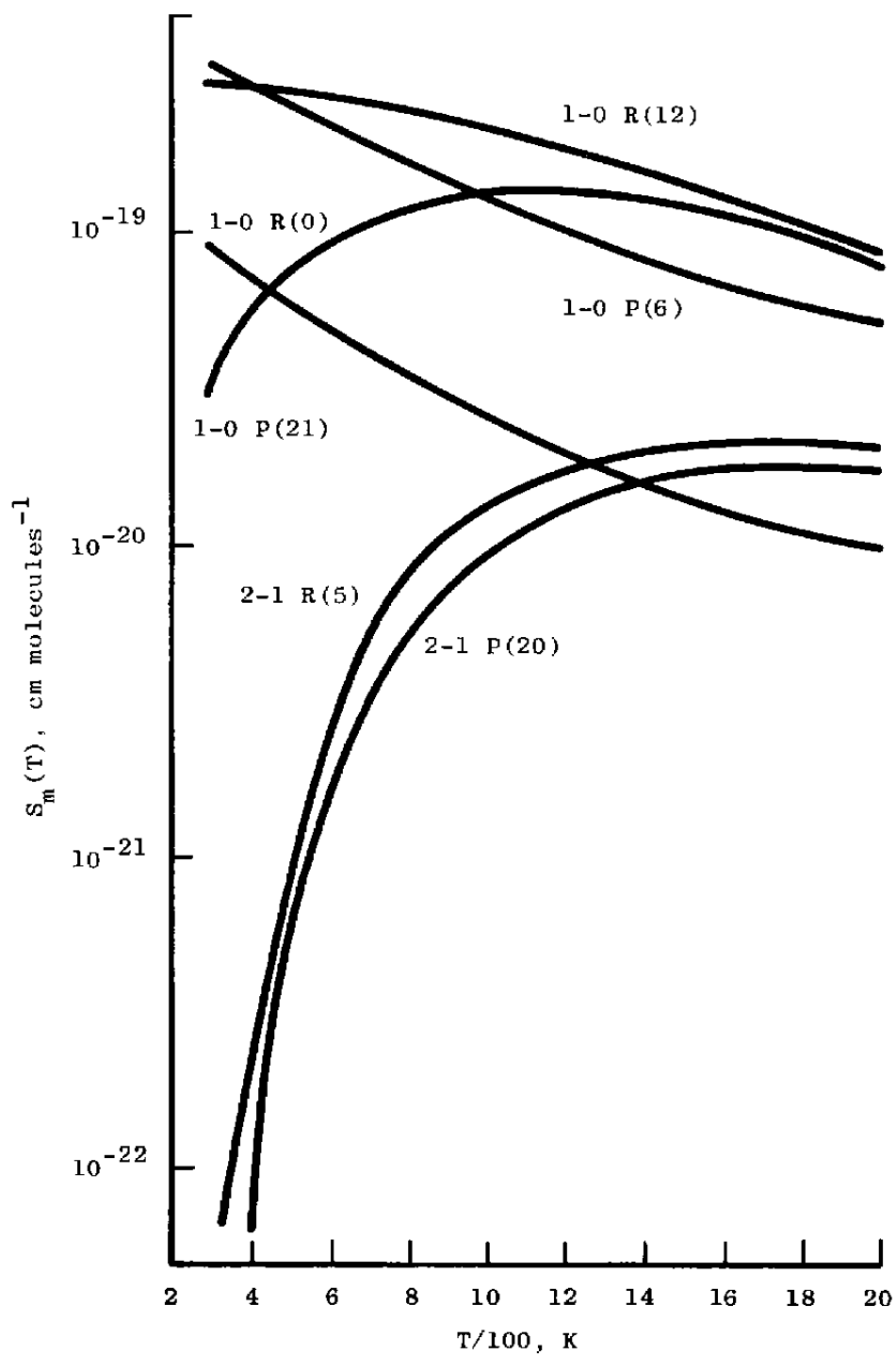


Figure 1. Typical line strength temperature dependencies.

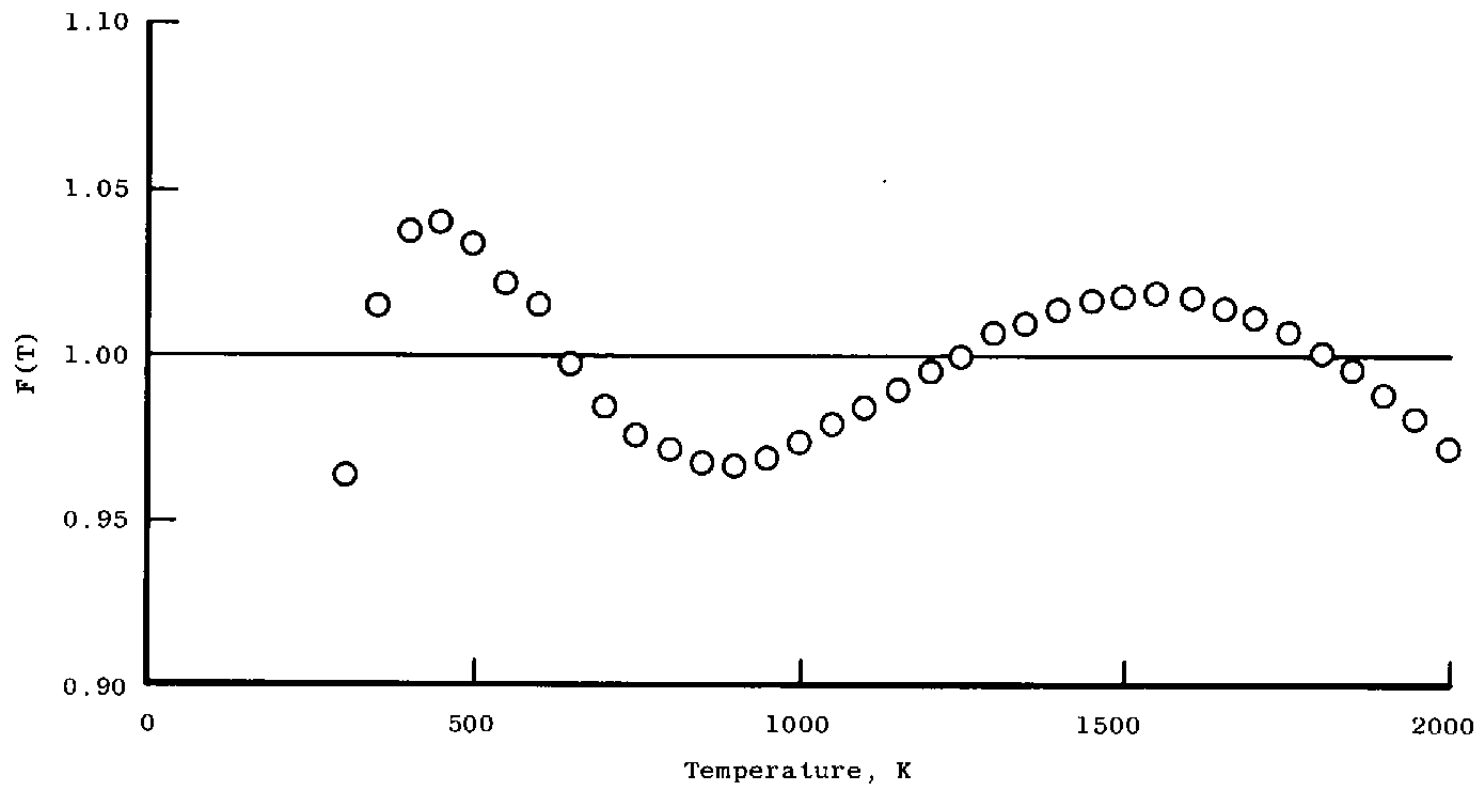


Figure 2. Temperature-minimized line strength function: 1-0 R(12), 2-1 R(19) combination.

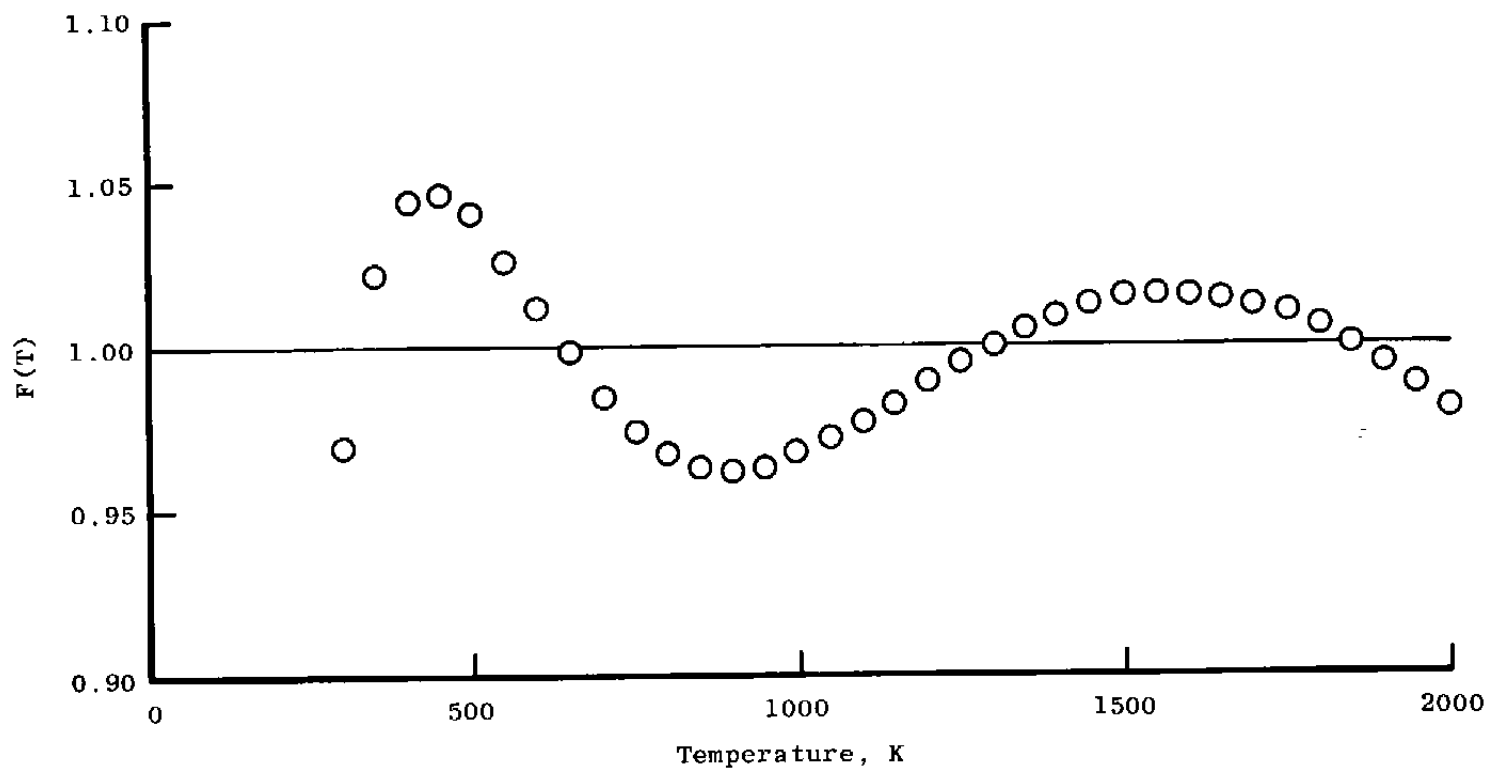


Figure 3. Temperature-minimized line strength function: 1-0 R(12), 2-1 R(20) combination.

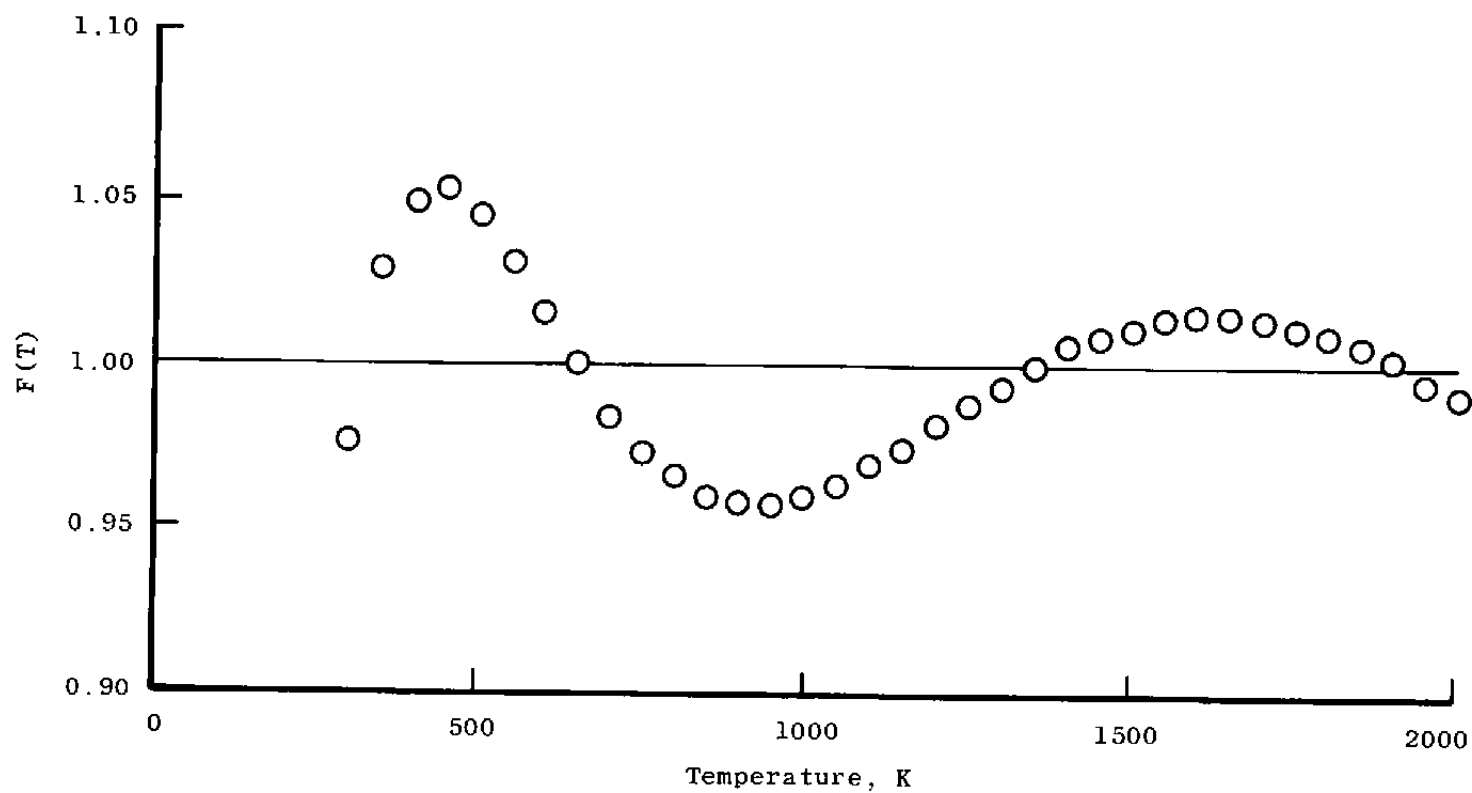


Figure 4. Temperature-minimized line strength function: 1-0 R(12), 2-1 R(21) combination.

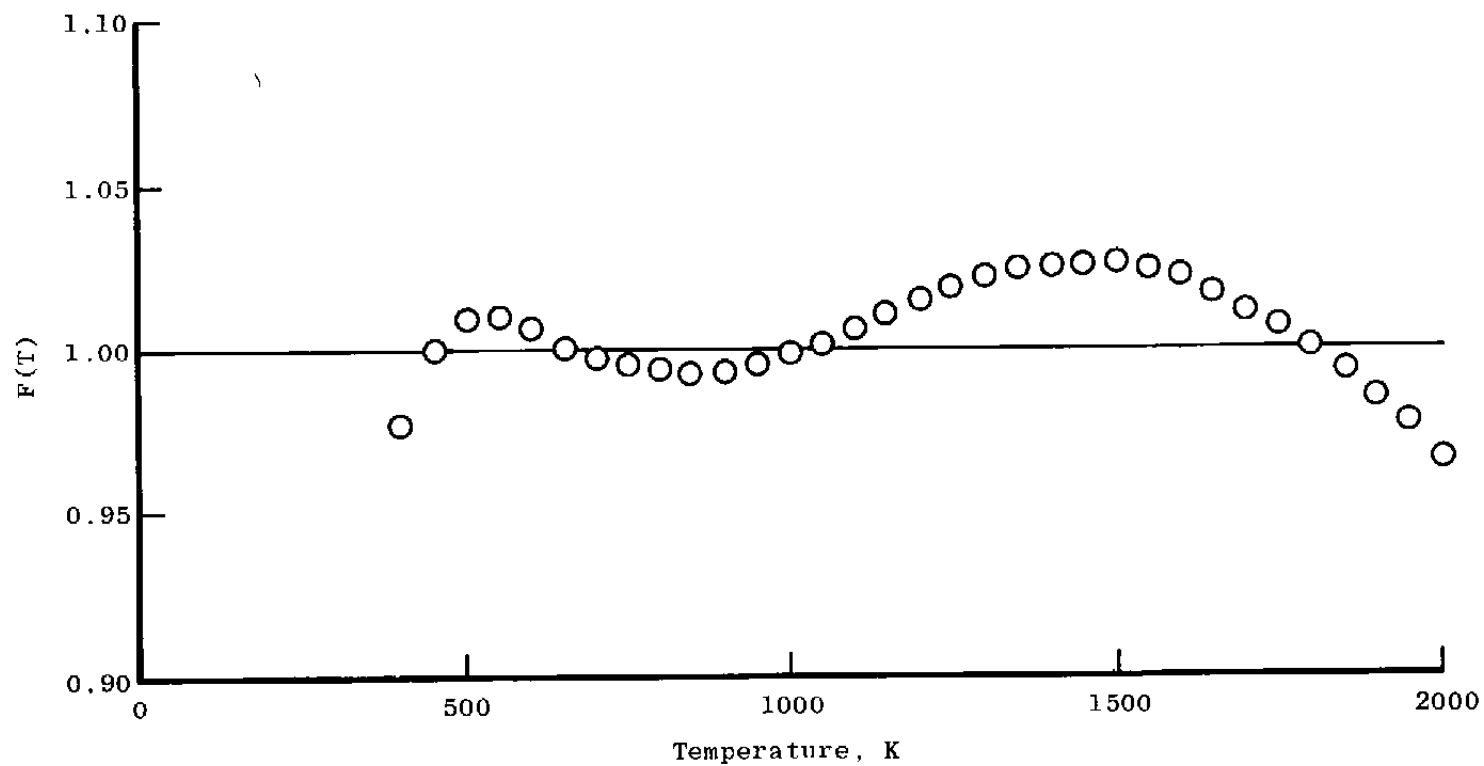


Figure 5. Temperature-minimized line strength function: 1-0 R(12), 2-1 R(19) combination.

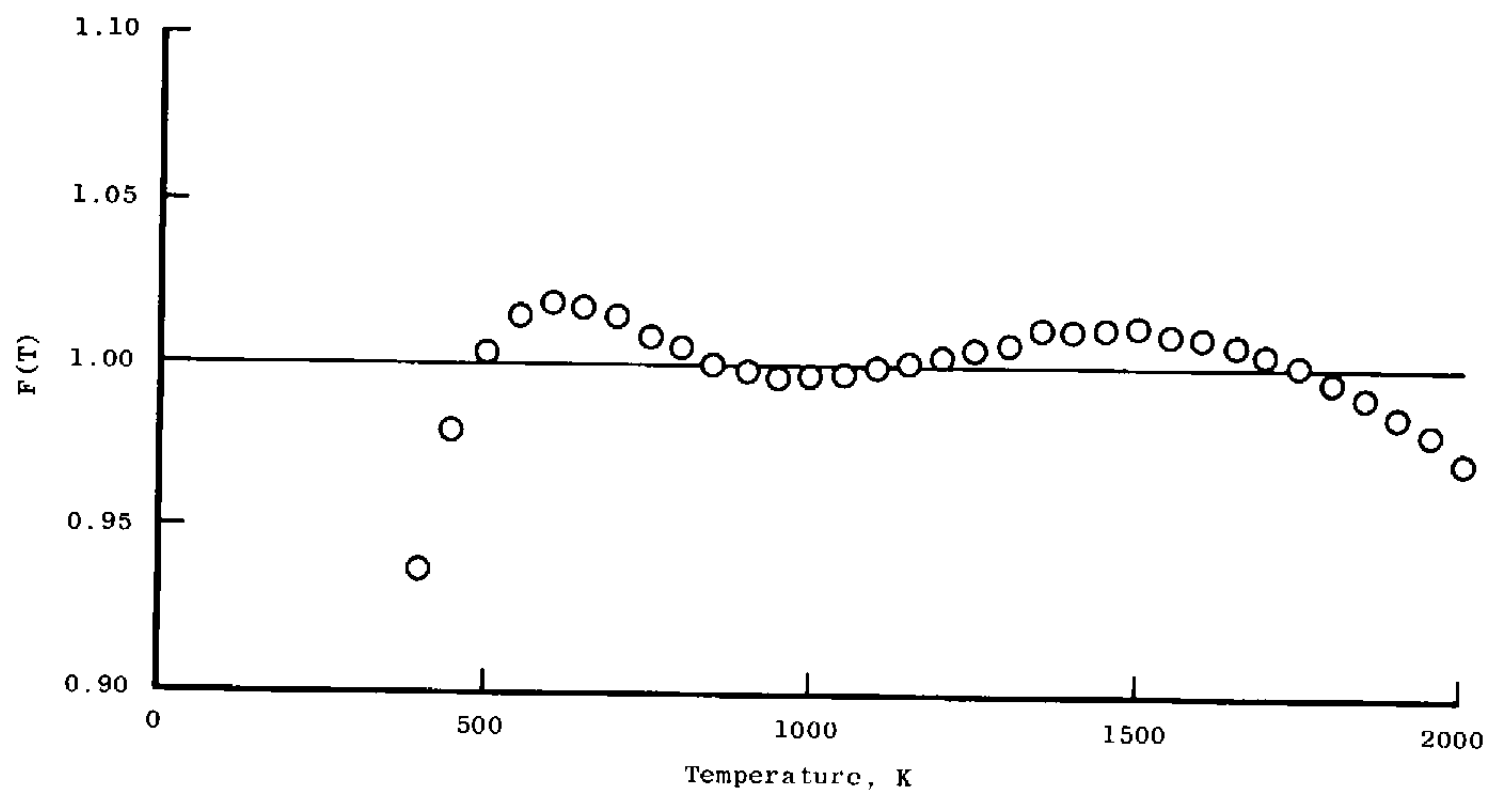


Figure 6. Temperature-minimized line strength function: 1-0 R(14), 2-1 R(23) combination.



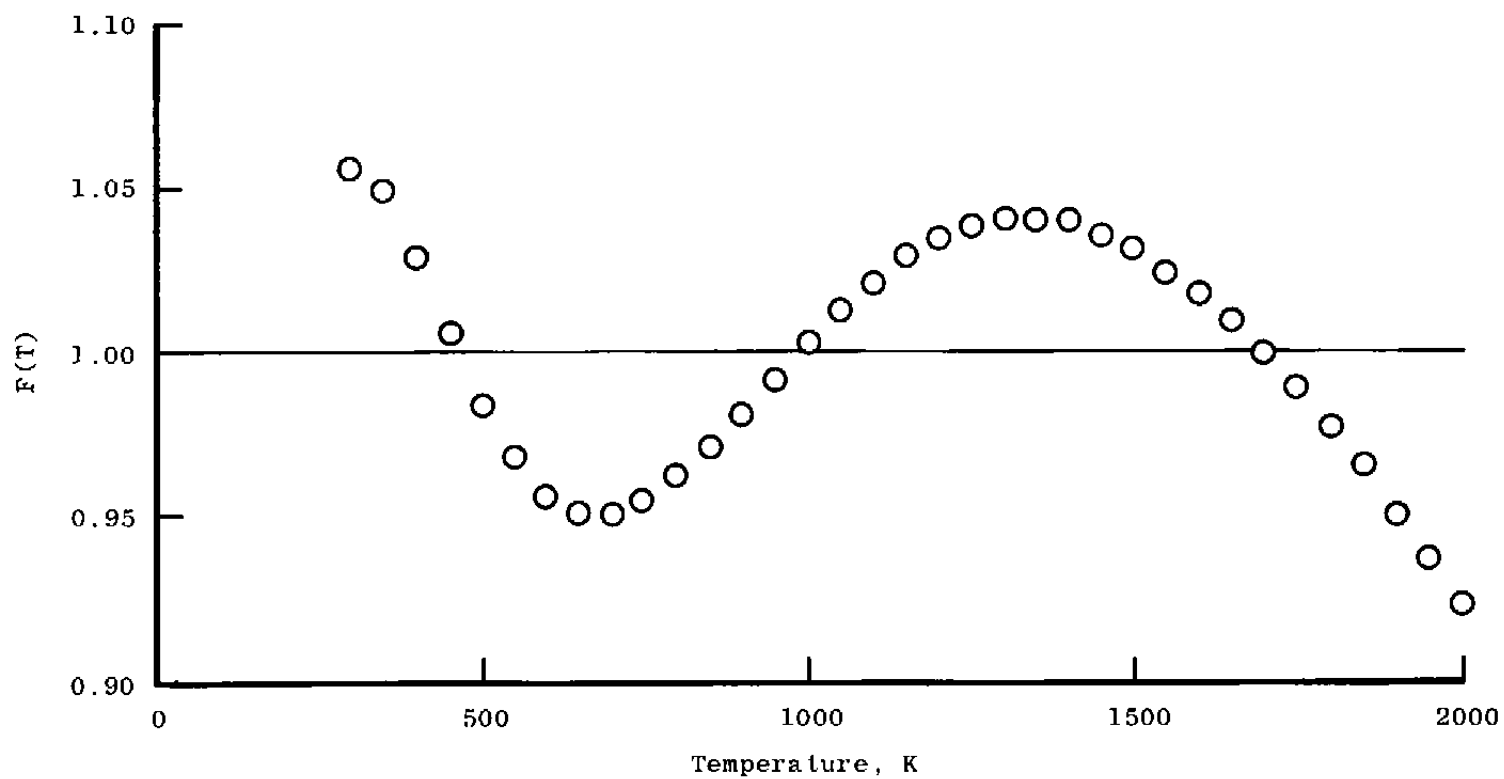


Figure 7. Temperature-minimized line strength function: 1-0 P(10), 2-1 P(6) combination.

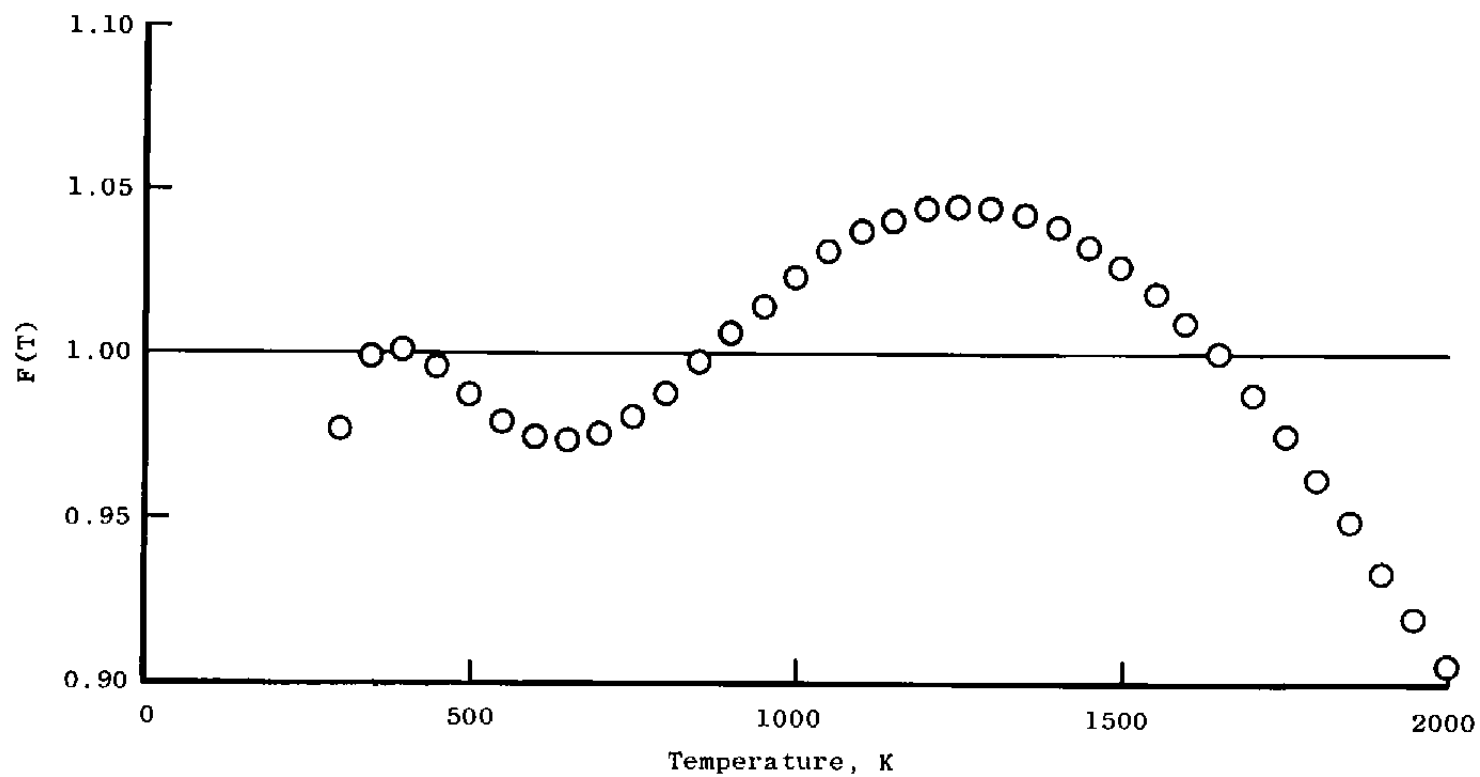


Figure 8. Temperature-minimized line strength function: 1-0 P(11), 2-1 P(5) combination.

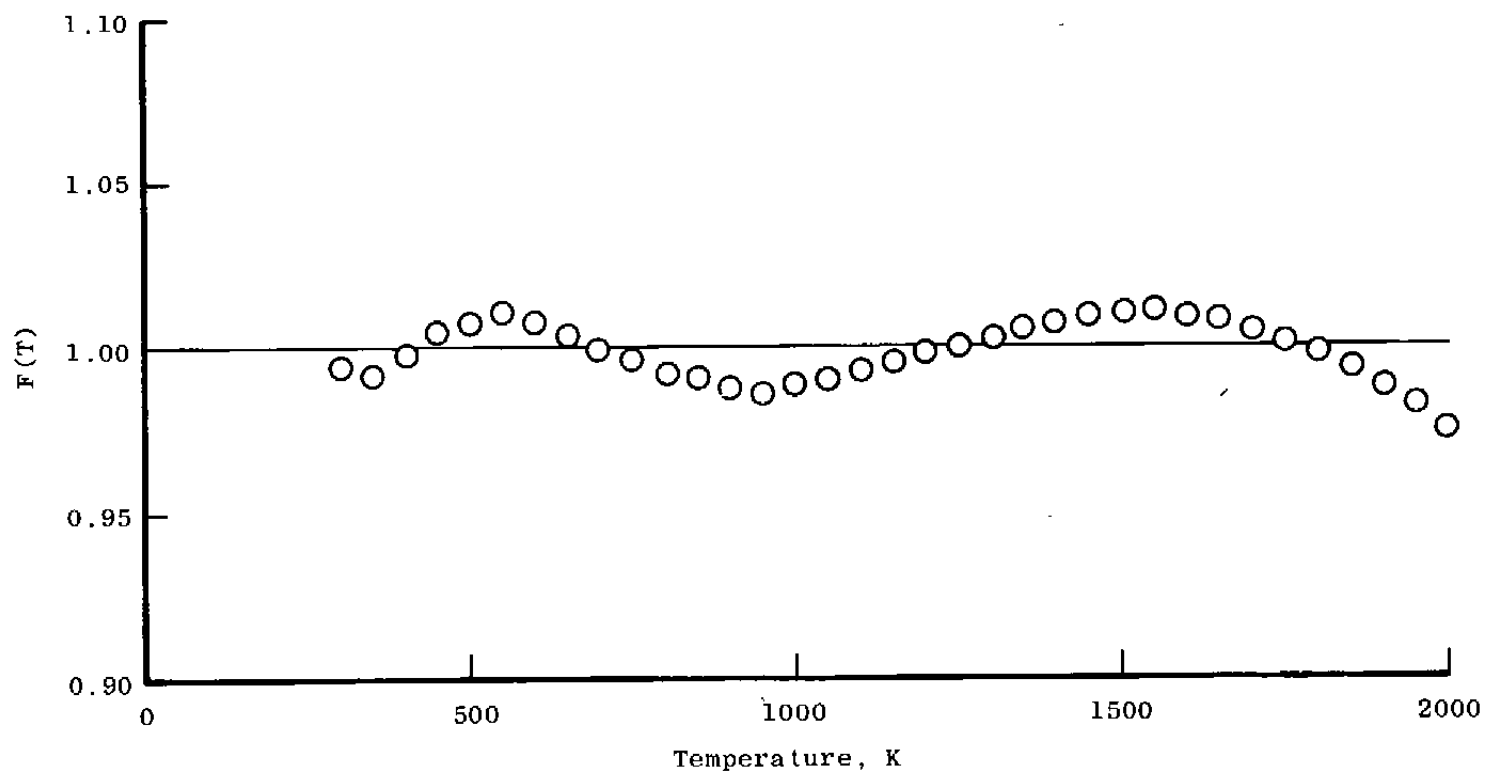


Figure 9. Temperature-minimized line strength function: 1-0 R(0), 1-0 R(17), 2-1 R(24) combination.

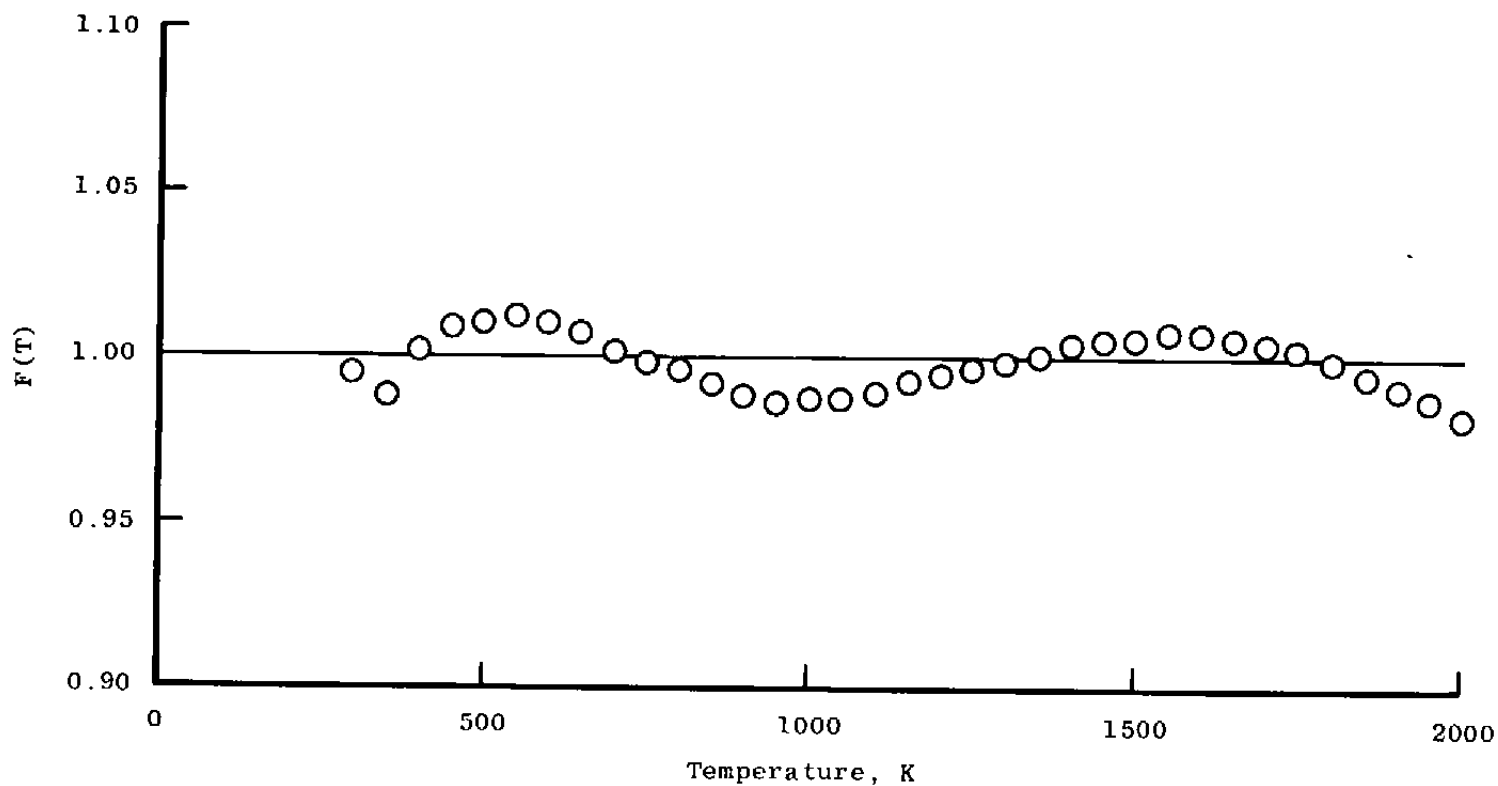


Figure 10. Temperature-minimized line strength function: 1-0 R(3), 1-0 R(18), 2-1 R(27) combination.

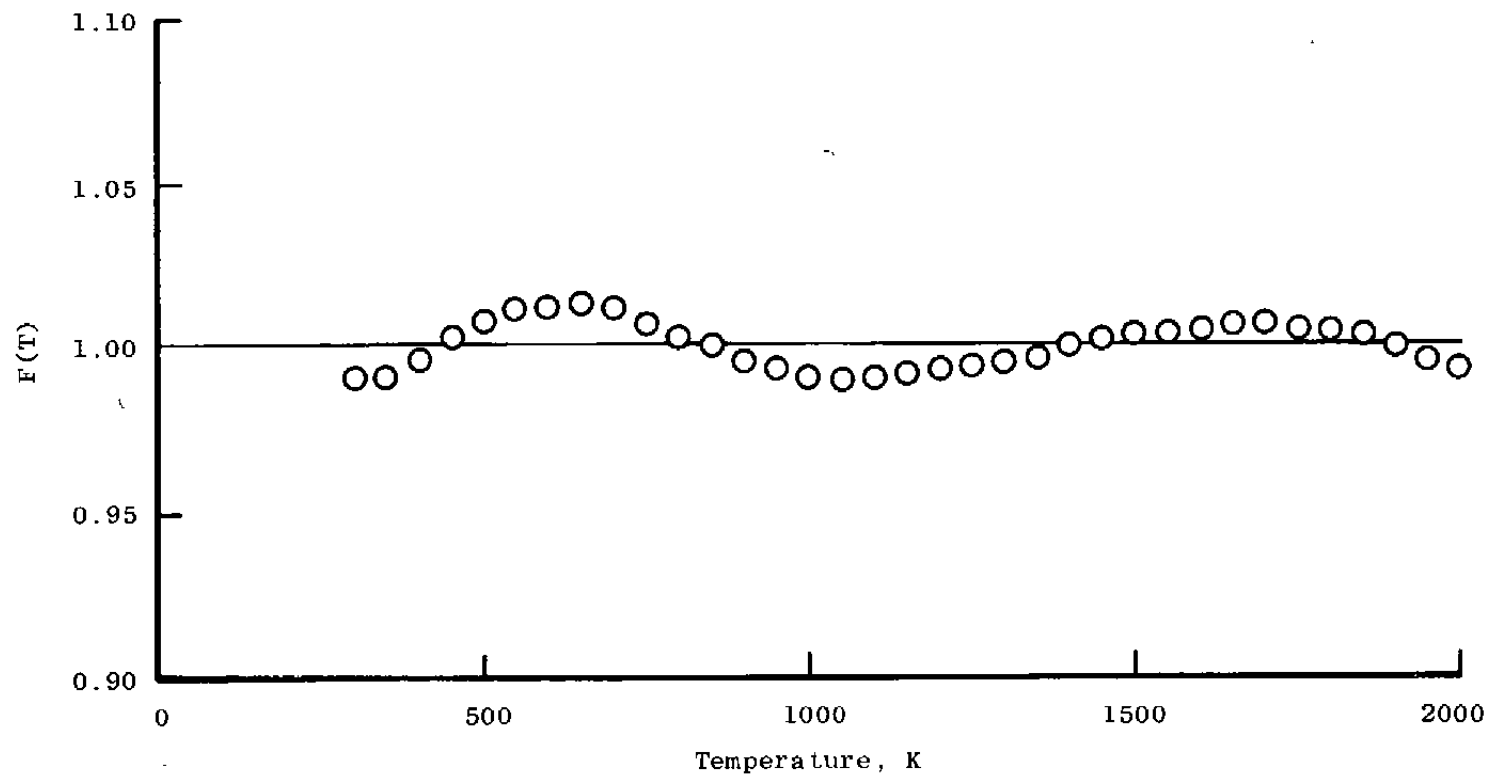


Figure 11. Temperature-minimized line strength function: 1-0 R(7), 1-0 R(20), 2-1 R(29) combination.

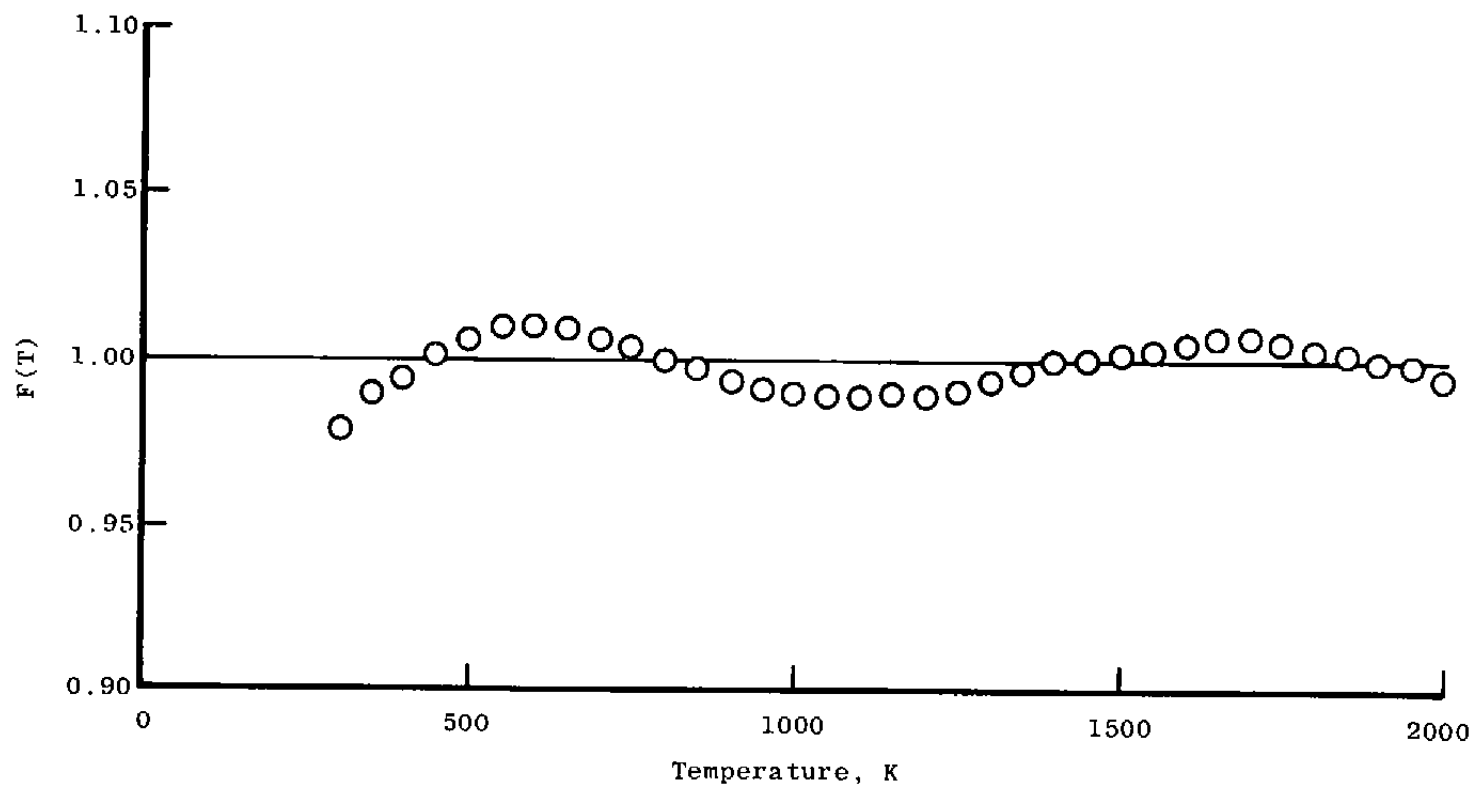


Figure 12. Temperature-minimized line strength function: 1-0 R(9), 1-0 R(22), 2-1 R(30) combination.

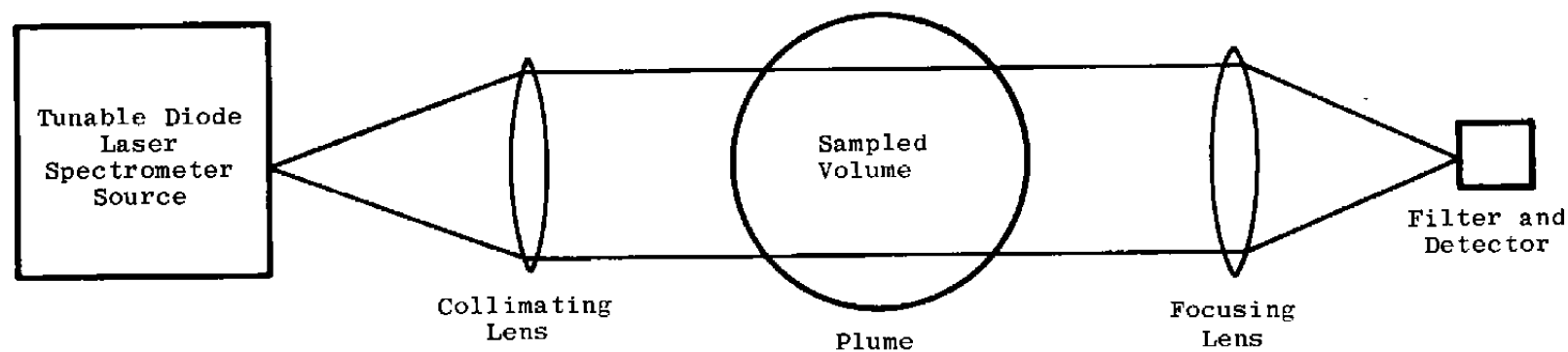


Figure 13. Schematic of an exhaust plume average species number density measurement system.

Table 1. Summary of Temperature-Minimization Calculations

Figure Number	Temperature Range, K	Lines	Line Positions, $\text{cm}^{-1}$	$S_m(298.15)$ , $\text{cm mol}^{-1}$	$a_i \times 10^{-18}$
2	300 to 2000	1-0 R(12)	2190.02	$2.93 \times 10^{-19}$	3.42
		2-1 R(19)	2185.45	$3.80 \times 10^{-24}$	4.96
3	300 to 2000	1-0 R(12)	2190.02	$2.93 \times 10^{-19}$	3.45
		2-1 R(20)	2188.49	$2.77 \times 10^{-24}$	4.84
4	300 to 2000	1-0 R(12)	2190.02	$2.93 \times 10^{-19}$	3.47
		2-1 R(21)	2191.50	$1.98 \times 10^{-24}$	4.75
5	400 to 2000	1-0 R(13)	2193.36	$2.49 \times 10^{-19}$	3.50
		2-1 R(19)	2185.45	$3.80 \times 10^{-24}$	4.65
6	400 to 2000	1-0 R(14)	2196.66	$2.06 \times 10^{-19}$	3.70
		2-1 R(23)	2197.39	$9.51 \times 10^{-25}$	4.02
7	300 to 2000	1-0 P(10)	2103.27	$3.32 \times 10^{-19}$	3.46
		2-1 P(6)	2093.41	$2.44 \times 10^{-23}$	15.21
8	300 to 2000	1-0 P(11)	2099.08	$2.97 \times 10^{-19}$	3.50
		2-1 P(5)	2097.39	$2.27 \times 10^{-23}$	17.04
9	300 to 2000	1-0 R(0)	2147.08	$9.40 \times 10^{-20}$	8.52
		1-0 R(17)	2206.35	$1.02 \times 10^{-19}$	2.93
		2-1 R(24)	2200.28	$6.39 \times 10^{-25}$	3.79
10	300 to 2000	1-0 R(3)	2158.30	$3.38 \times 10^{-19}$	2.57
		1-0 R(18)	2209.51	$7.71 \times 10^{-20}$	2.96
		2-1 R(27)	2208.72	$1.72 \times 10^{-25}$	3.54
11	300 to 2000	1-0 R(7)	2172.76	$4.53 \times 10^{-19}$	2.15
		2-0 R(20)	2215.70	$4.17 \times 10^{-20}$	2.64
		2-1 R(29)	2214.16	$6.50 \times 10^{-26}$	3.34
12	300 to 2000	1-0 R(9)	2179.77	$4.14 \times 10^{-19}$	2.44
		1-0 R(22)	2221.75	$2.06 \times 10^{-20}$	2.32
		2-1 R(30)	2216.81	$3.91 \times 10^{-26}$	3.16